

The superconducting phase of Calcium under the pressure at 200 GPa: the strong-coupling description

R. Szczęśniak, A.P. Durajski

*Institute of Physics, Częstochowa University of Technology,
Al. Armii Krajowej 19, 42-200 Częstochowa, Poland**

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The thermodynamic parameters of the superconducting state in Calcium under the pressure at 200 GPa have been determined. The numerical analysis by using the Eliashberg equations in the mixed representation has been conducted. It has been stated, that the critical temperature (T_C) decreases from 36.15 K to 20.79 K dependently on the assumed value of the Coulomb pseudopotential ($\mu^* \in (0.1, 0.3)$). Next, the order parameter near the temperature of zero Kelvin ($\Delta(0)$) has been obtained. It has been proven, that the dimensionless ratio $2\Delta(0)/k_B T_C$ decreases from 4.25 to 3.90 together with the growth of μ^* . Finally, the ratio of the electron effective mass to the electron bare mass (m_e^*/m_e) has been calculated. It has been shown, that m_e^*/m_e takes the high value in the whole range of the superconducting phase's existence, and its maximum is equal to 2.23 for $T = T_C$.

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I. INTRODUCTION

In Calcium under the influence of the high pressure (p), the whole sequence of the structural phase transitions is induced.

In the normal conditions Calcium crystallizes in the fcc structure (Ca-I). For the pressure at 20 GPa, the transition to the structure bcc (Ca-II) has been observed. The next change appears for the pressure's value at 32 GPa, above which the structure sc (Ca-III) is realized [1]. In 2005 Yabuuchi *et al.* have found another two crystal phases: Ca-IV and Ca-V [2], [3]. It has been stated, that the phase Ca-IV is stable from 119 GPa; above the pressure of 143 GPa appears Ca-V. The phases Ca-IV and Ca-V have been identified experimentally by Fujihisa *et al.* [4]. The following assignment has been proposed: Ca-IV with the $P4_12_12$ structure and Ca-V with the $Cmca$ structure. In the range of pressures from 158 GPa to 207 GPa, Nakamoto *et al.* have discovered the phase Ca-VI (the $Pnma$ structure) [5]. Above, Sakata *et al.* have reported existence of the host-guest structure (Ca-VII) [6].

The results achieved by using the *ab initio* methods only partially agree with the experimental scheme of the structural phase transitions. In particular, Ishikawa *et al.* and Arapan *et al.* have reproduced the appearance of the low pressure structures (fcc and bcc) [7], [8]. However, the values of the pressure, at which the structural transitions appear, differ from the experimental values. Above the pressure at 36 GPa, the sequences of the successive structural transitions, explicitly differ between them-

selves and do not reproduce the experimental scheme. In the context of the essential theoretical results, we have mentioned the paper of Yao *et al.* [9]. The authors were the ones who identified the phase Ca-IV as the structure $Pnma$ and the phase Ca-V as $Cmca$.

In the range of the very high pressures (from 135 GPa to 495 GPa), the calculations of Ishikawa *et al.* suggest the stability of the structure $I4/mcm(00\gamma)$. However, the similar value of the enthalpy has the $Pnma$ structure (at last for the lower pressures from the discussed range) [7]. It has to be underlined, that the stability of the $Pnma$ structure from 158 GPa to 180 GPa is suggested by the paper of Aftabuzzaman and Islam [10]. Also, the results presented by Yin *et al.* prove especially low values of the $Pnma$ enthalpy ($p \in (135, 220)$ GPa) [11].

Besides the very complex sequence of the structural transitions, which to the present day has not been finally determined, Calcium is being characterized with the interesting superconducting properties. In particular, the very high values of the critical temperature among the simple metals.

The possibility of the superconducting state's existence in Calcium has been reported in 1981 by Dunn and Bundy [12]. In 1996, the definite experiment has been performed by Okada *et al.* [13]. After ten years, the studies of Okada have been repeated by Yabuuchi *et al.* [14]. The researchers determined the dependence of the critical temperature on the pressure, at the same time ascertaining, that T_C appreciably exceeds the values determined by Okada.

According to Yabuuchi *et al.*, the superconducting state in Calcium is induced in the following crystal phases: Ca-III, Ca-IV and Ca-V [14]. In particular, in the phase Ca-III the critical temperature quickly increases together with the pressure's growth (from about 3 K for

*Electronic address: adurajski@wip.pcz.pl

$p = 58$ GPa to 23 K for $p = 113$ GPa). In the phase Ca-IV, the critical temperature is the subject of the saturation, while in the phase Ca-V it takes the value of 25 K at 161 GPa. In 2011, Sakata *et al.* have reported $T_C = 29$ K for $p = 216$ GPa [6]; the highest observed value of the critical temperature among all elements.

The above experimental results inspired us to estimate the basic thermodynamic parameters of the Ca superconducting state in the range of the very high pressures. In particular, we assume: $p = 200$ GPa. The calculations will be conducted in the framework of the Eliashberg formalism, with the aid of the Eliashberg function calculated by Yin *et al.* in the paper [11] (*Pnma* structure).

II. THE ELIASHBERG EQUATIONS

The Eliashberg equations have been derived in order to quantitative description of the superconducting state for the intermediate and strong coupling between electrons and phonons.

In appliance to the approach used in the BCS theory [15], the Eliashberg equations enable the detailed consideration of the complicated form of the electron-phonon interaction. In the result, the Eliashberg set allows exactly to estimate the value of the critical temperature, the order parameter and the electron effective mass, if the value of the Coulomb pseudopotential is known.

The Eliashberg equations can be written on the imaginary axis, on the real axis or in the mixed representation [16]. From the numerical point of view, the most convenient in application is the mixed representation, because the rather simple algorithms can be used [17], [18].

The Eliashberg equations in the mixed representation take the form [19]:

$$\begin{aligned} \phi(\omega) = & \frac{\pi}{\beta} \sum_{m=-M}^M \frac{\lambda(\omega - i\omega_m) - \mu^* \theta(\omega_c - |\omega_m|)}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} \phi_m \quad (1) \\ & + i\pi \int_0^{+\infty} d\omega' \alpha^2 F(\omega') [N(\omega') + f(\omega' - \omega)] \\ & \times K(\omega, -\omega') \phi(\omega - \omega') \\ & + i\pi \int_0^{+\infty} d\omega' \alpha^2 F(\omega') [N(\omega') + f(\omega' + \omega)] \\ & \times K(\omega, \omega') \phi(\omega + \omega'), \end{aligned}$$

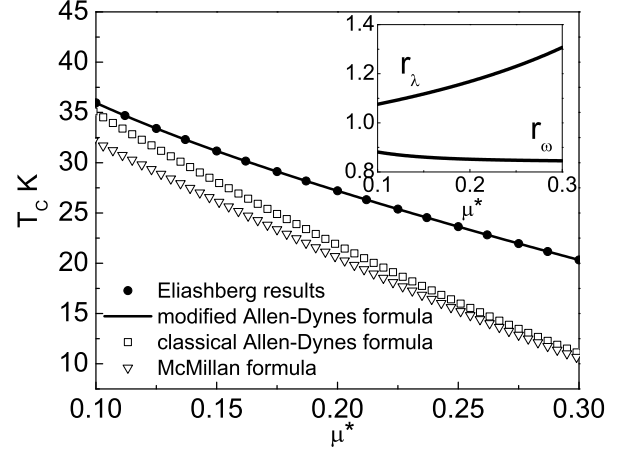


FIG. 1: The dependence of the critical temperature on the Coulomb pseudopotential. In the inset, we have shown the ratios r_ω and r_λ as the function of the Coulomb pseudopotential.

and

$$\begin{aligned} Z(\omega) = & 1 + \frac{i\pi}{\omega\beta} \sum_{m=-M}^M \frac{\lambda(\omega - i\omega_m) \omega_m}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}} Z_m \quad (2) \\ & + \frac{i\pi}{\omega} \int_0^{+\infty} d\omega' \alpha^2 F(\omega') [N(\omega') + f(\omega' - \omega)] \\ & \times K(\omega, -\omega') (\omega - \omega') Z(\omega - \omega') \\ & + \frac{i\pi}{\omega} \int_0^{+\infty} d\omega' \alpha^2 F(\omega') [N(\omega') + f(\omega' + \omega)] \\ & \times K(\omega, \omega') (\omega + \omega') Z(\omega + \omega'), \end{aligned}$$

where: $K(\omega, \omega') \equiv \frac{1}{\sqrt{(\omega + \omega')^2 Z^2(\omega + \omega') - \phi^2(\omega + \omega')}}.$

The symbols: $\phi(\omega)$, $Z(\omega)$, ($\phi_n \equiv \phi(i\omega_n)$ and $Z_n \equiv Z(i\omega_n)$) represent the order parameter function and the wave function renormalization factor on the real (imaginary) axis, respectively; ω_n is the n -th Matsubara frequency: $\omega_n \equiv (\pi/\beta)(2n - 1)$. The inverse temperature is given by: $\beta \equiv (k_B T)^{-1}$, and k_B is the Boltzmann constant. The order parameter is defined as: $\Delta \equiv \phi/Z$.

In the framework of the Eliashberg formalism, the pairing kernel for the electron-phonon interaction is determined in the following way: $\lambda(z) \equiv 2 \int_0^{\Omega_{\max}} d\Omega \frac{\Omega}{\Omega^2 - z^2} \alpha^2 F(\Omega)$. The symbol $\alpha^2 F(\Omega)$ denotes the Eliashberg function [11]; the value of the maximum phonon frequency (Ω_{\max}) is equal to 78.11 meV.

The depairing Coulomb interaction is modeled parametrically with the aid of the Coulomb pseudopotential μ^* . Symbol θ denotes the Heaviside unit function and ω_c is the cut-off frequency ($\omega_c = 3\Omega_{\max}$). The Bose-Einstein and Fermi-Dirac functions are represented by $N(\omega)$ and $f(\omega)$, respectively.

The Eliashberg equations have been solved numerically

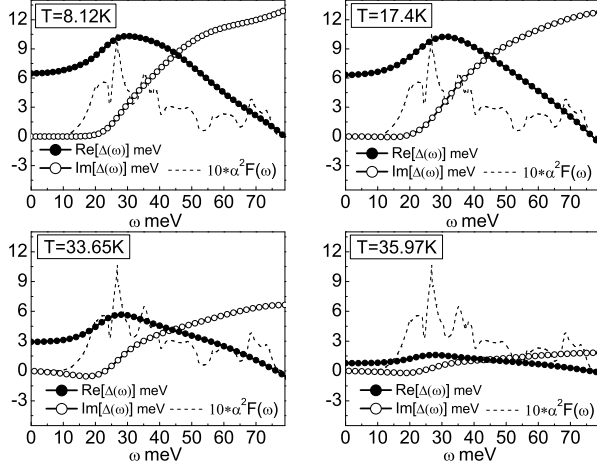


FIG. 2: The real and imaginary part of the order parameter on the real axis for selected values of the temperature. The rescaled Eliashberg function has been also plotted.

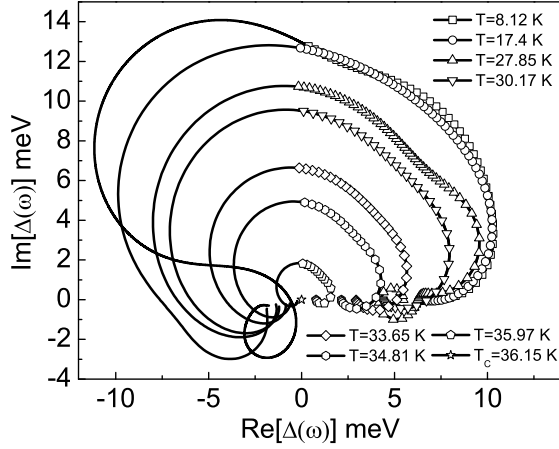


FIG. 3: The form of the order parameter on the complex plane for selected values of the temperature. The results obtained for $\omega \in \langle 0, \Omega_{\max} \rangle$ by using the lines with empty symbols have been denoted; the solid lines represent results achieved for $\omega \in (\Omega_{\max}, \omega_c)$.

for 2201 Matsubara frequencies ($M = 1100$). The stability of the solutions has been achieved for the temperatures greater or equal to $T_0 = 8.12$ K. Additional information on the means of the analysis of the Eliashberg equations, the reader can find in the papers: [20], [21], and [22].

III. THE RESULTS

In the Eliashberg formalism, the critical temperature depends on the form of the Eliashberg function and the value of the Coulomb pseudopotential. In the opposition to the Eliashberg function, the parameter μ^* is really

hard to calculate using the *ab initio* methods. According to the above, we have taken into consideration the very wide range of the Coulomb pseudopotential's values e.g. $\mu^* \in \langle 0.1, 0.3 \rangle$. The significant suggestion, that not only low values of μ^* should be considered, are the results obtained for Calcium under pressure at 120 GPa, where $\mu^* = 0.215$ has been noted [22].

In Fig. 1, we have presented the critical temperature as the function of μ^* . The exact results obtained by using the Eliashberg equations are represented by the filled circles. It can be easily noticed, that together with increase of the parameter μ^* , the critical temperature decreases from the value of 36.15 K to 20.79 K. The above result proves, that even for the very large value of Coulomb pseudopotential, T_C achieves high values. We notice, that the range of the calculated values of T_C agrees qualitatively with the recently experimental data. In particular, Sakata *et al.* have shown that: $T_C = 29$ K for $p = 216$ GPa [6].

From the mathematical point of view the determination of the critical temperature with the aid of the Eliashberg equations is the complicated and time consuming issue. For that reason, we have presented the analytical formula, which exactly reproduces the numerical results in opposition to the classical expressions of Allen-Dynes or McMillan (see Fig. 1) [23], [24]. In order to do that, the Allen-Dynes formula with the newly fitted parameters has been used. We notice, that the fitted parameters have been determined on the basis of the 250 exact values of T_C (μ^*), and the least squares method. The result takes the form:

$$k_B T_C = f_1 f_2 \frac{\omega_{\ln}}{1.45} \exp \left[\frac{-1.03 (1 + \lambda)}{\lambda - \mu^* (1 + 0.06\lambda)} \right], \quad (3)$$

where the strong-coupling correction function (f_1) and the shape correction function (f_2) are given by the expressions: $f_1 \equiv \left[1 + \left(\frac{\lambda}{\Lambda_1} \right)^{\frac{3}{2}} \right]^{\frac{1}{3}}$ and $f_2 \equiv 1 + \frac{(\frac{\sqrt{\omega_2}}{\omega_{\ln}} - 1) \lambda^2}{\lambda^2 + \Lambda_2^2}$. The symbol λ denotes the electron-phonon coupling constant: $\lambda \equiv 2 \int_0^{\Omega_{\max}} d\Omega \frac{\alpha^2 F(\Omega)}{\Omega}$. The quantity ω_2 represents the second moment of the normalized weight function: $\omega_2 \equiv \frac{2}{\lambda} \int_0^{\Omega_{\max}} d\Omega \alpha^2 F(\Omega) \Omega$, and ω_{\ln} is called the logarithmic phonon frequency: $\omega_{\ln} \equiv \exp \left[\frac{2}{\lambda} \int_0^{\Omega_{\max}} d\Omega \frac{\alpha^2 F(\Omega)}{\Omega} \ln(\Omega) \right]$. In particular, for Calcium under the pressure at 200 GPa, the following values have been obtained: $\lambda = 1.23$, $\sqrt{\omega_2} = 29.98$ meV, and $\omega_{\ln} = 35.92$ meV. The functions Λ_1 and Λ_2 are defined as: $\Lambda_1 \equiv 2.6 (1 + 1.8\mu^*)$, and $\Lambda_2 \equiv 0.092 (1 - 150\mu^*) \left(\frac{\sqrt{\omega_2}}{\omega_{\ln}} \right)$.

The modified Allen-Dynes (mAD) formula possesses the fitted parameters, which are sharply different than the parameters included in the classical expression derived by Allen and Dynes (AD). The above result is connected with the fact, that the effective phonon frequency ($[\omega_{\text{eff}}]_{\text{AD}}$) and the effective coupling constant ($[\lambda_{\text{eff}}]_{\text{AD}}$) in classical Allen-Dynes formula well reconstruct the exact

value of T_C only for low values of the Coulomb pseudopotential ($\mu^* \rightarrow 0.1$). We notice, that the parameters ω_{eff} and λ_{eff} have been defined as: $k_B T_C = \omega_{\text{eff}} e^{-\frac{1}{\lambda_{\text{eff}}}}$. In the case of high values of μ^* , the classical parameterization overestimates the effective phonon frequency and much underestimates the effective coupling constant. The inset in the Fig. 1 presents the detailed dependences of the ratios $r_\omega \equiv [\omega_{\text{eff}}]_{\text{mAD}} / [\omega_{\text{eff}}]_{\text{AD}}$ and $r_\lambda \equiv [\lambda_{\text{eff}}]_{\text{mAD}} / [\lambda_{\text{eff}}]_{\text{AD}}$ on μ^* .

Finally, we notice that the classical expressions for T_C should be applied with the great carefulness, because even for low-temperature superconductors like Al or Pb, the difference between analytical and numerical Eliashberg results are noticeable [25].

In Fig. 2, we have presented the order parameter on the real axis for the selected values of the temperature and $\mu^* = 0.1$. We see, that for the low frequencies, the non-zero value is taken only by the real part of $\Delta(\omega)$. The obtained result proves, that in the considered range of frequencies the damping effects not exist [20]. For the higher values of the frequency ($\omega \sim 30$ meV) one can observe the maximum of $\text{Re}[\Delta(\omega)]$, which is induced by the area of the characteristic peaks in the Eliashberg function. From the physical point of view, the above fact indicates that the relevant growth of $\text{Re}[\Delta(\omega)]$ exists in the frequencies' area in which the electron-phonon coupling is exceptionally strong. Additionally, it should be marked out, that in the considered case the function $\text{Im}[\Delta(\omega)]$ increases monotonically together with the growth of ω . For the frequencies higher than ~ 30 meV, the real part of the order parameter decreases together with the growth of ω due to the drop of the Eliashberg function's value. In the last step let us turn our attention to the fact, that the growth of the Coulomb pseudopotential causes only the significant decrease of the order parameter while the shapes of $\text{Re}[\Delta(\omega)]$ and $\text{Im}[\Delta(\omega)]$ remain very similar.

In Fig. 3, we have plotted the order parameter on the complex plane. It can be easily noticed, that the values of $\Delta(\omega)$ make the characteristic spirals with the radius that decreases together with the growth of the temperature. The obtained result allows to characterize the effective electron-electron interaction. In particular, for $\text{Re}[\Delta(\omega)] > 0$ the effective interaction is pairing, whereas for $\text{Re}[\Delta(\omega)] < 0$ the depairing processes are in major. In the case when $\mu^* = 0.1$, the area of the frequencies in which the electron-electron interaction is attractive covers the range of the frequencies for which the Eliashberg function is definite. As the result of the μ^* increasing, the frequencies that correspond to the positive value of $\text{Re}[\Delta(\omega)]$ become narrow from the side of Ω_{max} ; however such effect is not exceptionally strong.

On the basis of the obtained results, we have calculated the physical value of the order parameter for the given temperature ($\Delta(T)$). In order to achieve that, one should use the following equation [16]: $\Delta(T) = \text{Re}[\Delta(\omega = \Delta(T))]$. From the physical point of view, the most interesting is the value of the order parameter close to the temperature of zero Kelvin ($\Delta(0) \simeq \Delta(T_0)$),

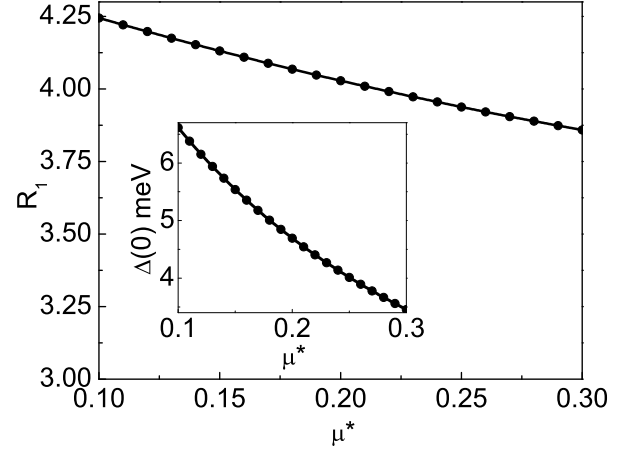


FIG. 4: The dependence of the ratio R_1 on the value of the Coulomb pseudopotential. In the inset, the influence of μ^* on the value of the parameter $\Delta(0)$ has been presented.

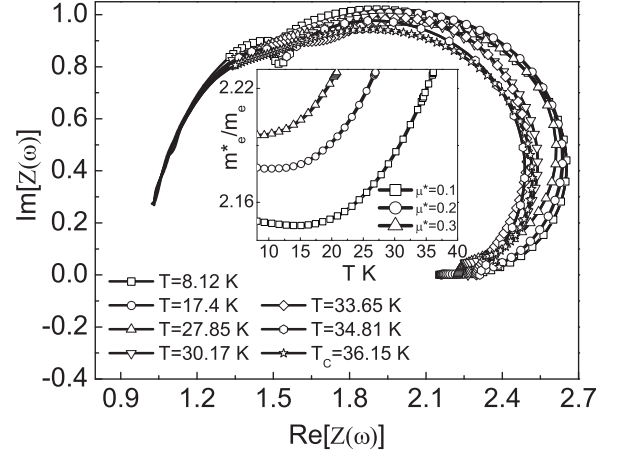


FIG. 5: The form of the wave function renormalization factor on the complex plane for selected values of the temperature and $\mu^* = 0.1$. The results for $\omega \in \langle 0, \Omega_{\text{max}} \rangle$ by using the lines with empty symbols have been denoted; the solid lines represent the results for $\omega \in (\Omega_{\text{max}}, \omega_c)$. In the inset, we have presented the dependence of the ratio m_e^*/m_e on the temperature for selected values of the Coulomb pseudopotential.

because it allows to determine the characteristic ratio: $R_1 \equiv 2\Delta(0)/k_B T_C$. Let us notice, that in the framework of the BCS theory, the parameter R_1 takes the universal value equal to 3.53 [15].

In Fig. 4, we have presented the results achieved for Calcium, where the dependence of R_1 on the Coulomb pseudopotential has been plotted. We see, that the parameter R_1 is always bigger than $[R_1]_{\text{BCS}}$ and only slightly decreases together with the increase of μ^* (from 4.25 to 3.90). We notice, that the weak dependence of the ratio R_1 on μ^* is connected with the fact that the Coulomb pseudopotential in the comparable way lowers the value of $\Delta(0)$ and T_C . In particular, the open de-

pendence of $\Delta(0)$ on μ^* in the Fig's. 4 inset have been plotted, whereas the shape of the function $T_C(\mu^*)$ in Fig. 1 has been shown.

From the physical point of view, the high values of the ratio $[R_1]_{Ca}$ are the consequence of the existence of the strong-coupling and retardation effects, which are omitted by the simple BCS theory. In the framework of the Eliashberg approach, these effects can be characterized with the aid of the parameter $k_B T_C / \omega_{ln}$. In the weak-coupling limit, one can assume: $[k_B T_C / \omega_{ln}]_{BCS} = 0$. In the case of Calcium, we have obtained: $[k_B T_C / \omega_{ln}]_{\mu^*=0.1} \simeq 0.104$, and $[k_B T_C / \omega_{ln}]_{\mu^*=0.3} \simeq 0.059$. Above results clearly differ from the BCS approximation.

Finally, we notice that the exact value of μ^* for Calcium under the pressure at 200 GPa is unknown. However, on the basis of the modified Allen-Dynes formula and the experimental result presented recently by Sakata *et al.* ($T_C = 29$ K for $p = 216$ GPa) [6], one can obtain $[\mu^*]_{p=216\text{GPa}} \simeq 0.177$. If we assume that: $[\mu^*]_{p=200\text{GPa}} \sim [\mu^*]_{p=216\text{GPa}}$, the value of $[R_1]_{p=200\text{GPa}}$ equals about 4.0.

In the Eliashberg formalism the dependence of the electron effective mass (m_e^*) on the temperature is determined by the wave function renormalization factor. In particular, we have: $m_e^* = \text{Re}[Z(0)] m_e$, where m_e denotes the electron bare mass.

In Fig. 5, we have plotted the values of $Z(\omega)$ on the complex plane in order to investigate the influence of the temperature on the form of the wave function renormalization factor. On the basis of the obtained results, it has been stated, that the wave function renormalization factor weakly depends on the temperature in comparison with the order parameter. We notice, that the increasing value of the Coulomb pseudopotential does not change significantly the course of $Z(\omega)$.

In the Fig's. 5 inset, the dependence of the ratio m_e^*/m_e on the temperature for the selected values of the Coulomb pseudopotential has been shown. The achieved results prove, that the electron effective mass takes the very high value in the whole range of the superconducting phase's existence, and m_e^* has the maximum for $T = T_C$, where $m_e^*/m_e = 2.23$.

IV. SUMMARY

In the paper, we have obtained the basic thermodynamic parameters of the superconducting state in Cal-

cium under the pressure at 200 GPa. It has been stated, that dependently on the assumed value of the Coulomb pseudopotential ($\mu^* \in \langle 0.1, 0.3 \rangle$), the critical temperature decreases in the range from 36.15 K to 20.79 K. This result is in agreement with the recently experimental data obtained by Sakata *et al.* ($T_C = 29$ K for $p = 216$ GPa) [6].

Next, the dependence of the order parameter $\Delta(0)$ on μ^* has been determined. The results allowed to determine the value of the ratio R_1 . We have affirmed, that the parameter R_1 is essentially larger than $[R_1]_{BCS}$, and slightly decreases with the growth of the Coulomb pseudopotential from 4.25 to 3.90.

In the last step, the electron effective mass has been determined. We have shown, that m_e^* is large and takes its maximum for $T = T_C$. In particular: $[m_e^*]_{\max} = 2.23 m_e$.

In the paper, the detailed characteristic of the order parameter on the real axis has been also presented. It has been proven, that the courses of $\text{Re}[\Delta(\omega)]$ and $\text{Im}[\Delta(\omega)]$ are clearly correlated with the shape of the Eliashberg function. Additionally, the plot of the order parameter on the complex plane enabled the conclusion, that for $\mu^* = 0.1$, the effective electron-electron interaction is attractive in the frequencies' range, where the Eliashberg function is definite. The increase of μ^* causes only the insignificant narrowing of the frequencies' range from the side of Ω_{\max} .

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